LETTER REPORT

TITLE: Review of "Measurements of Thermal Neutrons in the Subsurface," by M. W. Kuhn, S. N. Davis, H. W. Bentley, and R. Zito (1984). Geophysical Research Letters 11, 607-610.

AUTHOR: G. D. O'Kelley

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PROJECT MANAGER: G. K. Jacobs

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SUMMARY

Dating of groundwater by tracers such as ^{36}Cl may be sensitive to interferences from thermal-neutron production in the subsurface. Thermal neutrons are produced in the subsurface mainly by (a) cosmic-ray interactions with geological strata, (b) alpha-particle reactions on light elements, and (c) spontaneous fission of ^{238}U. This paper presents the first data on absolute neutron production rates by modern instrumental methods. Field measurements in this study were carried out on geological formations with several different chemical compositions and from depths of 12 to 1300 m. At the shallow subsurface, up to 116 neutrons cm^{-3} $year^{-1}$ were detected, which probably included a substantial cosmic-ray component. In mines below 800 m depth, which were effectively free of cosmic-ray effects, measurements of neutron production rates ranged from 1.1 to 33 neutrons cm^{-3} $year^{-1}$. Calculations of neutron production rates were compared with the measurements. At depths below 800 m, the authors concluded that more thermal neutrons were detected than calculated; however, from the stated experimental errors it is not clear that the differences are significant.

REVIEW OF REPORT

A number of radionuclides, which are produced continuously by interactions between cosmic radiation and gases in the upper atmosphere or by interactions between cosmic radiation and elements located at or just below the earth's surface, are candidates for possible use in dating of groundwater. The natural concentrations of some of these nuclides, e.g., ^{3}H, ^{85}Kr, and ^{14}C, have been masked to varying degrees by man-made sources. Indeed, ^{85}Kr, with a half-life of only 10.7 years, is so completely masked by the vast amount of man-made ^{85}Kr that its natural concentrations in water prior to 1945 may never be determined (Davis and Bentley, 1982). Other nuclides such as ^{39}Ar, ^{32}Si, ^{81}Kr, and ^{36}Cl are not being produced by *artificial means at present. However, a complication in the use of ^{36}Cl, ^{39}Ar, and ^{81}Kr is that these nuclides can be produced below the surface of the earth by thermal-neutron capture on stable elements. In the case of ^{36}Cl (half-life = 301,000 years), which has received considerable attention as a tracer for groundwater dating, subsurface production by the natural neutron flux may limit the useful range of dating. Bentley (1978) estimated the normal ranges for subsurface production of ^{36}Cl and concluded that it becomes significant after about two half-lives and may dominate the concentrations of ^{36}Cl after four half-lives.*

Rates of production of thermal neutrons in the deep subsurface can be obtained by three independent methods: (1) direct measurements, as described in the subject; (2) measurements of the accumulated products of neutron capture; and (3) theoretical calculations based on chemical analyses of the subsurface material, together with neutron yields from (alpha,n) reactions on these elements, and from natural fission events. The purpose of this report by Kuhn, et al. was to report measured values of neutron production rates in the subsurface and to compare these measured values with theoretical calculations. It is especially important to assess the accuracy of theoretical calculations of neutron production rates, since in some environments, such as uranium or thorium ores, the subsurface neutron production may be roughly equivalent to the atmospheric production (Davis and Bentley, 1982).

Thermal neutrons in the subsurface originate from effects of cosmic radiation; from alpha-particle-induced nuclear reactions, chiefly in light elements; and from uranium fission. In the upper atmosphere, protons and other cosmic-ray primary particles interact with the atmospheric constituents to produce large numbers of secondary particles, which include a strong neutron component. These neutrons interact with a rather thin (few cm) layer of exposed surface. However, the muons of the cosmic-ray secondary particles penetrate to great depths, interacting with geologic materials to produce neutrons in significant numbers, although the muon flux and the related neutron production rate diminish with increasing depth. The decay of naturally-occurring isotopes of thorium and uranium are the major sources of alpha particles which create neutrons through (alpha,n) reactions on light elements, although rare-earth radionuclides constitute minor sources of less-energetic alpha particles. Spontaneous fission, a relatively less probable decay mode of ^{238}U than alpha decay, is a significant source of neutrons in some geological settings. Calculations have indicated that, at high uranium concentrations, the neutron production rate is also increased by the fission of ^{235}U in natural uranium.

According to the authors, most state-of the-art calculations of neutron production rates based on the chemical composition of the surroundings make use of the following principal assumptions:

(1) The only sources of neutrons are (alpha,n) reactions and spontaneous fission of ^{238}U.

(2) The only sources of alpha particles are the decay of natural uranium, thorium, and their radioactive decay products.

(3) Nuclei of the elements present are distributed homogeneously.

(4) All neutrons thermalize rapidly in the subsurface without production of secondary alpha particles or neutrons.

The measurements were carried out using a ^3He proportional counter of conventional, low-background design. A preamplifier was mounted on the detector assembly to permit operation in bore holes up to depths of about 50 m. The instrument package was adequate, but not of a modern design. A measurement in a bore hole consisted of acquiring neutron events from the unshielded ^3He counter, then repeating the count with a cadmium shield in place to absorb the thermal neutrons. The events due to thermal neutrons were taken to be the difference between the unshielded and shielded counts. Where holes were not available, a half-shielded configuration was used, so that the neutron counting rate of the exposed geological surface could be determined. Details of the experimental method were reported in a thesis by Kuhn (1983). Experiments were carried out to demonstrate that gamma radiation had a negligible effect on the detector response. For safety of the electronic apparatus and for simplicity of data analysis, saturated portions of mine drifts and bore holes were avoided.

The counting data were converted to neutron production rates in units of neutrons cm^{-3} $year^{-1}$ by a straightforward integration procedure. The method is discussed in the paper and the mathematical formula is given there in equation (1). In Table 1 are shown the production rates as given in Table 1 of the paper, reordered according to depth of station and including only an abbreviated description of the location. The calculated values of the neutron production rates quoted in Table 1 were obtained by use of current methodology as described in several recent publications, whose references are given in the paper.

TABLE 1. Calculated and Measured Production Rates of Thermal Neutrons in the Subsurface (neutrons cm^{-3} $year^{-1}$).

Location	Calculated Production Rate	Measured Production Rate	Depth of Station (m)
Granite, drill hole	31 ± 3	116 ± 21	12
Alluvium, well	34 ± 5	53 ± 9.5	21
Altered, silicified limestone, mine	19 ± 2	42 ± 7.6	30
Altered granite, mine	28 ± 3	33 ± 5.9	900
Limestone, mine	5.8 ± 0.5	7.8 ± 1.4	1300
Conglomerate, mine	5.5 ± 0.6	1.1 ± 0.2	1300

The data reported in Table 1 are only the second set to be reported in the literature. Unfortunately, the first publication by Shmanin, et al. (1957) did not give enough experimental detail to derive production rates that could be compared with those of Table 1. Thus, the information in the present paper may be considered the only set of such data to be determined by modern instrumental methods.

With one exception, the calculated values of production rates in Table 1 tend to fall below the measured ones. Further, the differences *between these values tend to decrease with depth. As pointed out by the authors, the higher measured values at depths of 12 to 30 m are consistent with production of neutrons by cosmic-ray muons. Below 800 m the muon effect should be very small, and so the calculations should be expected to agree with the measurements, if the calculational method is correct. Although the authors appear to believe that the differences between calculated and measured values for altered granite at 900 m and for limestone at 1300 m constitute real, systematic discrepancies, this reviewer would argue that the calculated and measured values agree, since their stated errors essentially overlap and no details of the error analysis are given. The authors' claim that calculated values systematically underestimate neutron production rates also is in contradiction with the last line of Table 1, which shows that for one case the calculated neutron production rate is five times the measured value. It might be argued that, within experimental errors, the calculated and measured values agree at depths beyond those where significant effects of cosmic-ray muons are found, but that there may be some systematic experimental problem with the measurements associated with the conglomerate at 1300 m.*

EVALUATION OF REPORT

The publication represents a report of the first modern measurements of neutron production rates in the subsurface. It is, therefore, a useful reference on comparisons between calculated and measured values of production rates in geological environments with different chemical compositions. The paper is extensively annotated with references to relevant publications that are helpful to the reader. However, there are a few specific concerns related to the data and interpretations that are discussed below.

1. *Perhaps because the paper was published in a letters journal, it is summary in nature and is very short on experimental details and on discussion of error analysis. It would have been useful to have included in tabular form the uncorrected counting data together with the various numerical factors applied to achieve the final results. Such detail is common in low counting-rate experiments. For example, we do not know what the actual counting rates might have been; we are told only that counting periods were "..(generally measured in hours)..." No error analysis based on counting statistics was given; rather, all experimental errors for the production rates of Table 1 appear to be precisely 18%. Thus, there seems to have been an arbitrary assignment of error, perhaps based on conservative estimates of systematic factors in the experiments. If the authors were concerned about counting statistics, the counting intervals could have been increased to days instead of hours. With favorable counting statistics, it should have been possible to reduce over-all errors to considerably less than 18%.*

2. *The perceptive reader is bound to be curious as to why "...less than 0.4% of the thermal neutrons penetrate the shielding..." In shielding against thermal neutrons with cadmium, the amount of shielding can be made effectively 100%, because of the high cross section for cadmium and the*

exponential nature of the absorption process. Were there mechanical restrictions on the thickness of cadmium used or were there openings in the shield not properly protected? In the absence of an explanation, a reader might be inclined to question the use of an incompletely shielded detector.

3. *Although no information was given that would indicate that the electronic system was flawed, it seems that more modern electronic modules would have enhanced the stability and reliability of the measurements.*

4. *As indicated in the review above, the data on calculated vs. measured production rates as a function of chemical composition and depth do not support the authors' contention that calculated values fall consistently below measured values at depths beyond 100 m. The calculated and measured values agree within the stated errors in two of the three cases. To ascertain the extent of agreement or disagreement will require a more careful, quantitative analysis of all sources of error for both measurements and calculations.*

5. *When the authors had available a work area of low cosmic-ray background, such as a mine at a depth of 1300 m, it would have been very helpful to test the calculational methods by recording neutron counting rates in a detector imbedded in a pure matrix such as $CaCO_3$, then in a powder matrix of $CaCO_3$ containing an accurately known, homogeneous admixture of U_3O_8. Counting several such synthetic mixtures of pure components would validate the calculations in a direct way. Since ^{36}Cl dating may have to rely on calculated corrections of this sort, such direct checks on the calculations should be considered for future work.*

6. *Finally, it would have been more useful to have studied a broader range of chemical compositions at depths below several hundred meters. This is not easy work, but it would be of interest to measure a number of additional cases with greater precision and absolute accuracy than in these preliminary experiments.*

REFERENCES

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